The University Of Alabama In Huntsville

LOW LOSS WINDOW MATERIALS FOR CHEMICAL LASERS

by

James A. Harrington, PI

205-895-6404
Department of Physics
The University of Alabama in Huntsville
Huntsville, Alabama 35807

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The optical absorption has been measured at DF and HF wavelengths in a wide variety of materials which show promise for use as windows on high powered chemical lasers. These measurements, which were made using DF-HF chemical laser calorimetric techniques, will be discussed in terms of the

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individual hosts studied and collectively as well in order to obtain an overview of all the absorption data taken to date. Focusing on a compilation of data not only allows a quick determination of the lowest loss materials currently available but also enables one to draw several important conclusions regarding optical absorption in the 3 to 5 micron region. One significant but perplexing result is the fact that no total absorption coefficient measured in this region by us to date is less than 10^{-4} cm^{-1}. This is unusual because most materials studied have intrinsic levels far below this value and some of the same materials have measured absorptions less than 10^{-4} cm^{-1} at 5.25 and 1.06 microns. The reason for the higher absorptions in the DF-HF laser region is shown to be in part due to the higher surface absorption here than at other infrared laser frequencies. The identification and characterization of these surface absorption mechanisms (e.g. those arising from organic solvents used in surface cleaning and from mechanical polishing) will be discussed.

The specific materials studied in detail include ZnSe and SrF_2 from Raytheon, Ge and Si from Cal Tech, NaF and alkaline earth fluorides from NRL, CaF_2 from Hughes Research Labs, Yttralox from GE, and other miscellaneous alkali halides including NaCl, KCl, LiF, and KBr. Since surface absorption has been a prime concern, many of these samples had their surface and bulk absorption measured. ZnSe and SrF_2 showed a high surface absorption (10^{-3} cm^{-1} range) while the alkali halides and some CaF_2 exhibited little. At this point the lowest loss materials at 3.8 and 2.7 microns would be the alkaline earth fluorides and NaCl, ZnSe among semiconductors, and Yttralox among oxides.

PREFACE

The final technical report summarizes our study of the optical properties of low loss window materials for chemical lasers. The work described herein was performed during the period from 9 January 1974 to 10 November 1975. Participating in the research were, in addition to the principal investigator James A. Harrington, Don Gregory, James Rowe, Bill Otto, and Ben Ferguson.

SUMMARY

The increasing interest and importance of chemical lasers has led to a demand for materials studies at DF and HF chemical laser frequencies. The primary objective of this research program has been to investigate the optical absorption in a wide variety of low loss materials suitable for use as windows on high-powered chemical lasers. In order to accomplish this objective, the small optical absorption in a number of different state-of-the-art samples was measured using DF-HF chemical laser calorimetric techniques. These measurements, combined with a correlative study of surface absorption, have been compiled in order to bring together in a systematic fashion all the absorption data accumulated during the contract period. Such a compilation of data enables one to easily identify those materials which, up to the present time, hold the greatest promise, in terms of service and reliability, as chemical laser windows.

During the initial stages of the research program, while the small cw chemical laser was being constructed for calorimetric measurements, emphasis was placed on developing chemical polishes for the alkaline earth fluorides. Reasonably successful polishes were developed for CaF_2 and BaF_2 but no polish was found for SrF_2. The polishes that were developed will help to eliminate surface scratches and pits which should reduce surface absorption and scattering as well as minimize

laser induced surface damage from high-powered lasers.

Calorimetric measurements of the optical absorption coefficient at chemical laser frequencies are reported on a wide range of materials. These include the alkaline earth fluorides, alkali halides, zinc selenide and other semiconductors, various oxides, and some glasses. The absorption coefficient data also span a large range from 10^{-4} cm^{-1} to 10^{-1} cm^{-1}. This variation is due, in part, to the basic difference in the intrinsic absorption levels among the various hosts. For example, the oxide materials like sapphire have a much higher intrinsic or multiphonon absorption at the chemical laser frequencies than do, say, the alkaline earth fluorides. Hence, the absorption on oxide materials is ultimately limited by fundamental processes while absorption in most of the other materials studied is limited only by extrinsic processes (e.g. surface, impurity, mechanical defects) and thus presumably can be extremely small for the most technologically advanced materials.

The lowest absorbing materials naturally received the greatest attention. These materials, which for the most part were the alkaline earth fluorides and alkali halides, should in principle have the smallest absorption coefficient β because their absorption is entirely extrinsic at DF and HF wavelengths. One significant but perplexing result emerges from our analysis of the materials and this is that no total

absorption coefficient measured less than 10^{-4} cm^{-1}. This is unusual because some of the same materials have measured β's much less than 10^{-4} cm^{-1} at 10.6, 5.25, and 1.06 μm. The reason for the higher absorptions in the DF-HF region may be due, in part, to the higher surface absorption here than at the other infrared laser frequencies. Even this source of absorption, strong as it may be in some cases, is still felt not to be the primary reason for this discrepancy. Other explanations such as impurity absorption and absorption by mechanical imperfections have also been considered, but, as yet, no completely satisfactory source or sources of absorption have been advanced to explain this result.

The compilation of absorption data indicate that, for all the materials studied to date, the most promising window materials in terms of β only would be the alkaline earth fluorides and the alkali halides among ionic materials and ZnSe or silicon among semiconductors. It is expected, however, that improvements in materials technology and in surface polishing and cleaning methods will no doubt be forthcoming. We can anticipate, therefore, that these and other materials will exhibit even lower β's in the future.

I. INTRODUCTION

Over the period of performance of the contract, a wide variety of materials were investigated for possible use as windows on high-powered chemical lasers. Our primary criteria for assessing the window potential for all materials studied was the amount of optical absorption in the material at DF and HF chemical laser frequencies. The optical absorption is an important parameter in judging the ultimate success and reliability of a particular host as a laser window because the figure of merit of a material, which can be written in terms of the power transmitted[1], varies inversely as the absorption coefficient β. The lowest absorbing materials, therefore, are naturally the best window candidates based on optical absorption alone.

The major portion of this research program has, therefore, centered around careful measurements of the optical absorption in many different hosts using chemical laser calorimetric techniques. As the results were accumulated and interpreted, several general trends in the optical properties of materials became evident. One of these was that as materials processing technology improved so did the optical absorption coefficient. This evolutionary process at the chemical laser frequencies is similar to that observed over the years at CO_2 frequencies. There is, however, one major difference between materials improvement at 10.6 and that at 3.8 μm: while many 10.6 μm

window materials have been improved to the point that β's are currently measured in the 10^{-5} cm^{-1} range, no 3.8 μm window material has yet exhibited an absorption below 10^{-4} cm^{-1}. In fact this discrepancy seems to be fundamental in the sense that no materials show absorptions less than 10^{-4} cm^{-1} at either chemical laser (DF or HF) wavelength, yet some of the same materials have measured β's in the 10^{-5} to 10^{-6} cm^{-1} range at 10.6, 5.3, and 1.06 μm.[2] This is unusual because most materials studied by us have intrinsic levels far below 10^{-4} cm^{-1} so that one would not expect a priori very low β's at, say, 5.3 or 1.06 μm and significantly higher β's at the chemical laser frequencies. The explanation for this perplexing result is not known for certain. It may be that extrinsic absorption mechanisms are responsible. At HF wavelengths, the presence of OH^- impurities can give rise to higher β's, but at DF wavelengths these ions should not significantly contribute to the absorption. Furthermore, it is difficult to postulate a bulk impurity which can contribute to the extrinsic absorption at 3.8 μm. Another extrinsic process which could be influential is surface absorption. It is well known that certain common organic solvents used in cleaning and polishing window materials have strong absorptions in the 2.7 to 3.8 μm region. Any organic residue may thus lead to a strong surface contribution to the total absorption. In principle, however, this contribution can be separated from the bulk absorption[3] in order to compare

only bulk β's at the infrared laser frequencies. When this is done the optical absorption at chemical laser wavelengths is still higher. Efforts are continuing under a companion program to hopefully resolve this unexpected and unexplained result.

II. EXPERIMENTAL RESULTS AND DISCUSSION

The experimental results of the absorption coefficient at chemical laser frequencies for almost all of the hosts measured to date have already been discussed and summarized in three previous semi-annual ARPA reports.[4] These results will not be repeated; only the recent data will be presented.

The most recent samples studied are alkali halides, alkaline earth fluorides and cast SrF_2 obtained from Raytheon. These results are summarized in Table I.

In the case of KCl and KBr these materials represent some of the best crystals grown to date by NRL. Even so the β's at DF and HF frequencies are still higher than expected. The cast SrF_2 from Raytheon is a representative result from five samples studied. These β's are low and represent some of the best SrF_2 we have studied to date. Finally, the two sapphire samples indicate again[4] that the value at DF frequencies of 2.3×10^{-3} cm^{-1} is essentially the intrinsic value for this host so that no lower value can be expected. The sapphire suppliers, Crystal Systems and Union Carbide, use quite

Table 1

Absorption Coefficients for Miscellaneous Hosts at DF and HF Wavelengths

Sample	Source	Wavelength (microns)	β_{total} (cm^{-1})
KCl Single Crystal	Adolf Meller	3.8	3.3×10^{-4}
		2.7	1.2×10^{-3}
KCl Single Crystal	NRL B-254	10.6	4.0×10^{-4}
KBr Single Crystal	NRL B-212	10.6	8.9×10^{-5}
		3.8	5.6×10^{-4}
		2.7	3.8×10^{-4}
SrF_2 Polycrystal	Raytheon VHP-400	3.8	3.5×10^{-4}
		2.7	3.3×10^{-3}
BaF_2 Single Crystal	Adolf Meller	3.8	2.0×10^{-4}
		2.7	1.4×10^{-3}
CaF_2 Single Crystal	Adolf Meller	3.8	2.1×10^{-3}
		2.7	2.2×10^{-3}
Al_2O_3 (Sapphire)	Crystal Systems	3.8	2.3×10^{-2}
		2.7	3.4×10^{-3}
Al_2O_3 (Sapphire)	Union Carbide	3.8	2.5×10^{-2}
		2.7	2.6×10^{-3}

different growth methods, yet, as one would expect, there is effectively no difference at DF wavelengths because both materials are intrinsic.

III. SUMMARY OF RESULTS

The data in Table 1 combined with all previous results[4] enable one to quickly determine the range of β's observed for any particular host studied (cf. the last semi-annual report, August, 1975 for a compilation of all data up to that time). It is expected that materials will continue to improve and thus these lists of data will be updated to show the corresponding changes in β.

It is possible to cautiously draw some conclusions about the best potential laser window materials. Caution is necessary because only the absorption coefficient is considered in drawing these conclusions. In practice one would also consider other physical parameters of the material such as those involved in the figure of merit. With these restrictions in mind one can say that the alkaline earth fluorides are one of the best window candidates at this time. Equally good are the alkali halides. The best oxide material appears to be Yttralox. Silicon and ZnSe are the most promising semiconductors. As mentioned, these recommendations would be tempered by other physical properties of the material as well as by the specific application. In any case, these choices will likely continue

to be those for the future as well. Hopefully, as crystal growth technology improves, these materials should improve to an even greater extent in the future.

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